On the resonant optical bistability condition.

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Abstract

We address a two level system in an environment interacting with the electromagnetic field in the dipole approximation. The resonant optical bistability induced by local-field effects is studied by considering the relationship between the population difference and the excitation field. The diversity of various systems is included by accounting for the system self-action via the surface part of the Green's dyadic in the general form. The bistability condition and the exact solution of the steady state optical Bloch equations at the absolute bistability threshold are derived analytically.

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Because of its underlying nature and possible applications in the field of all-optical processing, the optical bistability (OB) was a subject of an intense experimental and theoretical research.[1, 2] In the early studies the use of a saturable absorber to induce OB was suggested.[3–7] The phenomenon was demonstrated experimentally for a cell of sodium vapor enclosed in a Fabry Perot interferometer and excited by a cw dyelaser.[8] Later it was conjectured that the local-field corrections alone could give rise to the mirrorless OB. [9] This type of bistability was extensively studied[10–14] and observed experimentally.[15]

The practical interest in all-optical devices faded to some extent as their solid state counterparts proved to perform better in terms of the switching speed and device density. However, the OB and optical hysteresis remain of considerable interest from the fundamental standpoint as a clear manifestation of a nonlinear light-matter interaction. Some new types of bistability mechanisms were discussed recently.[16–19] Besides, the question of the OB and hysteresis has received a renewed attention in connection with novel hybrid 0D nanoscopic systems, e. g., an artificial molecules comprising a semiconductor quantum dot (SQD) and metal nanoparticles (see Refs. 20–23 and references therein).

In this paper we address only the mirrorless OB induced by the local-field effects on a two-level system (TLS) interacting with the electromagnetic field in the dipole approximation. The local-filed correction leads to a self-action of the system, which results in a nonlinear relation between the applied field and the one acting upon the system. This type of the OB mechanism can be relevant for a large variety of systems: dense 3D assemblies of two-level atoms [9, 14], optically dense thin films of TLS [24, 25] or films of linear molecular aggregates, [26–28] hybrid metal-semiconductor systems, [20–23] or a more general case of a TLS in an environment involving dielectric and conducting surfaces, such as, a stratified media, a microcavity or a nanostructure.

OB can occur within a range of internal system parameters and external field intensities; identifying these ranges is therefore an important problems and its analytical solution is desirable. To the best of the author's knowledge, so far, it has been solved exactly only for two particular cases. Thus, Friedberg et al obtained analytically the bistability condition for the limiting case in which the active mechanism of the feedback is the nonlinear Lorentz shift of the resonance in a 3D gas of two-level atoms, which resulted from the near-field corrections. [14] Ignoring these corrections, Zakharov and Manykin derived the exact bistability criterion in the other limit in which the self-action is due to the radiated secondary field in

a thin 2D film of TLS. [25] The case when both these fields contribute into the self-action has been studied only numerically so far. [29]

We consider the simplest optical TLS coupled to an environment. The self-action of the TLS due to the environment can be described by the surface part of the Green's dyadic evaluated at the position of the TLS. Therefore, in this case the nonlinearity is characterized by a complex number which should satisfy some condition for the OB to occur. Below, we derive such condition analytically.

Following Refs. 14, 25, 29 we address the OB condition by considering the relationship between the population difference and the excitation field within the framework of the Bloch equations for the 2 × 2 density matrix ρ_{mn} (m, n = 0, 1) of a TLS. In the rotating wave approximation these equations read:

$$\dot{Z} = -\gamma (Z+1) - \frac{1}{2} [\Omega P^* + \Omega^* P] ,$$
 (1a)

$$\dot{P} = -(\Gamma + i\,\Delta)\,P + \Omega\,Z\,\,,\tag{1b}$$

where $Z = \rho_{11} - \rho_{00}$ is the population difference between the excited and the ground state of the TLS, P is the amplitude of the off-diagonal density matrix element defined through $\rho_{10} = -(i/2)P \exp(-i\omega t)$, γ and Γ are the relaxation constants of the population and the dipole moment, respectively, $\Delta = \omega_0 - \omega$ is the detuning of the driving field frequency ω from the TLS resonance ω_0 , and $\Omega = \mu \mathbf{E}/\hbar$ is the total electric field \mathbf{E} (in frequency units) acting upon the system, while μ being the TLS optical transition dipole. The dipole moment of the system is $\mathbf{p} = -i P \mu$, where P is its complex amplitude.

To calculate the total field Ω the Maxwell's equations are to be solved for a particular geometry. However, for a single dipole in an environment the total field Ω can be represented in the following general form:

$$\Omega = \widetilde{\Omega}_0 + \Omega_2, \quad \Omega_2 = -i G P \tag{2}$$

where $\widetilde{\Omega}_0$ is the renormalized incident field and Ω_2 is the secondary dipole field. The latter field is due to the feedback of the environment and is related to the corresponding response function[30–32]. It is determined by the surface part of the Green's dyadic evaluated at the position of the dipole. The near-zone component of the secondary dipole field, governed by $\operatorname{Re}(G)$, can originate from the near-field corrections, [14] while the far-zone contribution, governed by $\operatorname{Im}(G)$, can be due to the radiation. [25] In hybrid nano-systems comprising a

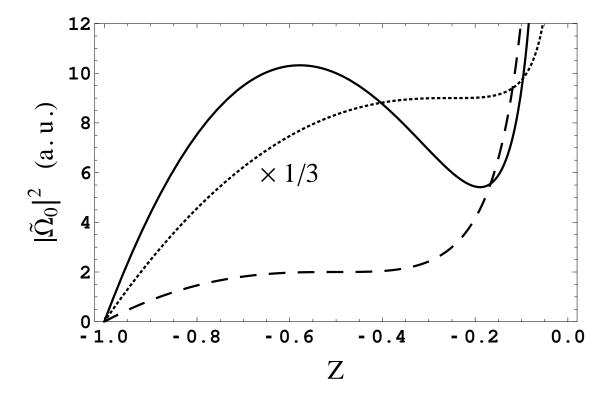


FIG. 1: The RHS of Eq. (3a) for $G=6, \Delta=1$ (solid line), $G=4, \Delta=1$ (dashed line) and $G=8i, \Delta=0$ (dotted line)

SQD, the feedback is provided by the secondary reflected field of the optical transition dipole moment. Thus, independently of its physical mechanism, the self-action is determined by the single complex valued parameter G. The total field Ω acting upon the TLS depends therefore on its state. Together with the nonlinearity of the TLS that can give rise to a variety of effects, such as the OB.

Under the steady-state conditions Eqs. (1) read:

$$\frac{|\widetilde{\Omega}_0|^2}{\gamma \Gamma} = -\frac{Z+1}{Z} \frac{(\Gamma - G_{\rm I}Z)^2 + (\Delta + G_{\rm R}Z)^2}{\Gamma^2} , \qquad (3a)$$

$$P = \frac{Z \widetilde{\Omega}_0}{(\Gamma - G_{\rm I} Z) + i (\Delta + G_{\rm R} Z)} , \qquad (3b)$$

where $G_R = \text{Re}(G)$ and $G_I = \text{Im}(G)$. The above equation highlights a known result of cavity quantum electrodynamics: the real part of the Green's dyadic shift the frequency while the imaginary part renormalizes the decay rate.[30–32] Both effects can be active mechanisms of the OB.

Eq. (3a) is of the third order in Z and can therefore have three real roots for some values

of Δ , Γ , G_R , and G_I (Γ is used as a unit rate from now on). These three solutions are different when the right hand side (RHS) of Eq. (3a) has a minimum and a maximum (see the solid line in Fig. 1), corresponding to two real roots of the RHS derivative, which satisfy the following equation

$$2|G|^{2}Z^{3} + (|G|^{2} + 2G_{R}\Delta - 2G_{I})Z^{2} - (1 + \Delta^{2}) = 0.$$
(4)

A threshold for bistability occurs when Eq. (4) has a double root (merged extrema – see the dashed or dotted lines in Fig. 1). In this case the solutions of Eq. (3a) are:

$$Z_1 = Z_2 = -\frac{|G|^2 + 2(G_R\Delta - G_I)}{3|G|^2}, \quad Z_3 = -\frac{Z_1}{2}$$
 (5)

Using the above root $Z_{1,2}$ and the condition $-1 \le Z \le 0$ one obtains an important constraint for the detuning Δ :

$$G_{\rm R} \Delta - G_{\rm I} \le |G|^2 \tag{6}$$

The formal condition of the existence of the double root (5) is determined by the following equation on Δ :

$$(|G|^2 + 2G_R \Delta - 2G_I)^3 - 27|G|^4 (1 + \Delta^2) = 0,$$
(7)

which is cubic in Δ and can also have three real solutions. It can be shown that only two of them, say $\Delta_1 < \Delta_2$, satisfy the constraint (6) and yield the upper and lower threshold values of the population difference Z and the external field $\widetilde{\Omega}_0$. Therefore, Eq. (4) has three different real roots within a window of detunings $\Delta_1 \leq \Delta \leq \Delta_2$. The absolute bistability threshold occurs when the window shrinks into a point, i. e., when a degenerate root $\Delta_1 = \Delta_2$ of Eq. (7) appears. After some algebra, one can obtain the following alternative for the existence of such double root:

$$G_{\rm R}^4 + 2(G_{\rm I}^2 - 4G_{\rm I} - 8)G_{\rm R}^2 + G_{\rm I}^3(G_{\rm I} - 8) = 0$$
, (8a)

$$G_{\rm I} + 1 = 0$$
 . (8b)

Only two of the solutions of the biquadratic equation (8a) satisfy the constraint (6). Analyzing the roots of Eq. (4) in the vicinity of the solution to Eqs. (8b) and (7), one finds that the system can be bistable below the line given by $G_{\rm I} = -1$, while being critical (i. e.,

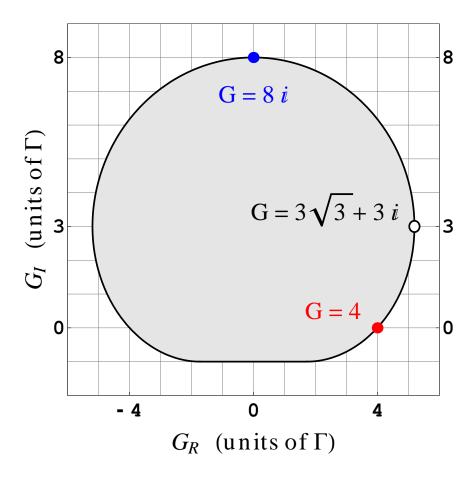


FIG. 2: The stability phase map: outside the gray area the system can be bistable. Solid line represents the phase boundary given by Eq. (9). See text for more details.

 $Z_{1,2}=-1$) on the line for $|G_{\rm R}|\leq\sqrt{3}$. Finally, the absolute bistability threshold is:

$$G_{\rm R}^2 = 8 + 4G_{\rm I} - G_{\rm I}^2 + 8\sqrt{1 + G_{\rm I}}, -1 < G_{\rm I} \le 8$$
 (9a)

$$-\sqrt{3} \le G_{\mathcal{R}} \le \sqrt{3}, \qquad G_{\mathcal{I}} = -1 \tag{9b}$$

For completeness, we provide also the expression for the corresponding detuning:

$$\Delta = \frac{|G|^4 - 4G_R^2 (G_I + 3) - 4G_I^2 (G_I - 1)}{|G|^4 + 8G_I [G_I^3 + G_R^2 (G_I + 2)]} G_R.$$
(10)

Equation (9) provides the condition of existence of three real roots of Eq. (3a). We studied the stability of these by analyzing Lyapunov exponents of Eqs. (1) in the vicinity of a stationary solution and calculated the stability phase map numerically. Figure 2 shows such phase diagram in the space of the feedback parameter G. Three different real solutions of Eq. (3a) exist within the white area, two of them being stable. The line dividing the two

phases represents the analytical condition Eq. (9) which gives also the bistability threshold. The general criterion Eq. (9) recovers both reported exact results: $G_{\rm I} > 8$ for $G_{\rm R} = 0$ [25] and $G_{\rm R} > 4$ for $G_{\rm I} = 0$ [14] (the corresponding threshold values of G are marked by full circles in Fig. 2). Note the mirror symmetry of the bistability map with respect to the line $G_{\rm R} = 0$, which reflects the invariance of Eq. (3a) under the simultaneous change of signs of $G_{\rm R}$ and Δ [29].

Eqs. (5) and (10) give the full solution of the steady state problem at a bistability threshold, while Eq. (9) is the bistability condition, which constitute the main result of the paper.

Finally, we note that novel hybrid plasmonic nano systems, such as a SQD and a metal nanoparticle complex (see Ref. 23 and references therein) or a SQD embedded in a stratified medium are excellent model systems with adjustable nonlinearity. The self-action field in these complexes is the secondary reflected field of the SQD optical transition dipole moment acting back upon the SQD. In the most general case such environment feedback results in a complex self-action parameter G which can be engineered by an appropriate choice of materials, geometry and/or external control parameters. Thus, if the SQD transition frequency is far from the plasmon resonance of the system then, typically, $|G_R| \gg |G_I|$ and the dominant mechanism of the bistability is the nonlinear Lorentz shift [14]. If the excitation frequency is near the surface plasmon resonance then the feedback can be almost purely imaginary: $|G_{\rm I}| \gg |G_{\rm R}|$ with $G_{\rm I} > 0$. Algebraically, this case is equivalent to the one of the radiation induced self-action [25]. A very interesting case is $G_{\rm I} < 0$ which can hardly be realized in the traditional extended 3D or 2D systems of two-level atoms. Nanoscopic hybrids are much more promising from this point of view due to their greater diversity. In the latter case various instabilities such as auto-oscillations can be expected under steadystate excitation. However, the detailed study of these instabilities goes beyond the scope of the present contribution and will be analyzed elsewhere.

Summarizing, we addressed the mirrorless resonant optical bistability of a two-level system in an environment by considering the relationship between the population difference and the intensity of the excitation field. The feedback of the environment was introduced in a general from via the surface part of the Green's dyadic. We derived the analytical bistability condition and obtained the exact steady state solution of the Maxwell-Bloch equations at the absolute bistability threshold. Our findings open the possibility to easily analyze diverse physical systems, determine experimental conditions (such as, the appropriate de-

tuning from the bare resonance and the intensity of the external field) necessary to observe the bistability and other nonlinear optical effects, as well as design and engineer new systems with desirable nonlinear optical properties.

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